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- (54) Improvements in or relating to polytetramethylene terephthalateshaped bodies
- (57) Unreinforced or reinforced polytetramethylene terephthalateshaped bodies is stabilised with triglycidyl isocyanurate and/or a

bisoxazoline, as well as optionally a phenolic or a phosphitic antioxidant. Shaped bodies can be produced from such stabilised polymer, especially by injection moulding, with reduction of the viscosity of the polymer and deterioration of the material properties of the pure polymer itself to any significant extent.

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Improvements in or relating to polytetramethylene terephthalate-shaped bodies

This invention relates to polytetramethylene terephthalate (PTMT) shaped bodies, which may be reinforced or unreinforced and which possess an enhanced property profile as well as improved 5 stability during working, and to a process for the production of such shaped bodies by co-extrusion.

Because of its good use properties and its ease of workability PTMT, as a thermoplastic polyester, has become of some significance as an injection moulding material. However a disadvantage of this polymer is the degradation it undergoes to products with strongly reduced viscosity numbers and deteriorated material properties as a result of the high working temperatures which need to be used 10 when producing shaped bodies from it by working in an extruder and then shaping it by means of for

This degradation is especially significant when PTMT is reinforced by addition of reinforcing fillers example injection moulding. or contains conventional flame protecting agents. The addition of impact resisting agents gives rise likewise to the degradation of PTMT and does not increase the impact and notched bar impact

This degradation is especially high and obviously to be avoided if reinforcing fillers, flame 15 resistance to the expected extent. protecting agents and impact resisting agents are used in combination in PTMT. For many uses PTMT has to be impact resistant at room temperatures and as much as possible at temperatures therebelow. Such behaviour should not be obtained to the detriment of the high rigidity and resistance to bending

20 It has already been proposed to improve the notched bar impact resistance of unreinforced PTMT 20 generally required of PTMT. by compounding it with polyethylene or ethylene/vinyl acetate copolymers (German Offenlegungsschrift 28 55 512), with polybutadiene onto which has been grafted styrene and methyl methacrylate (U.S. Patent 3,919,353), with a copolymer of butyl acrylate and tricyclodecenyl acrylate Onto which has been grafted styrene and acrylonitrile or a copolymer of butyl acrylate, butadiene and 25 methylvinyl ether onto which has been grafted styrene and acrylonitrile (German Auslegeschrift 24 44 584), with an ethylene/vinyl acetate copolymer on to which has been grafted acrylic acid or an ethylene/butyl acrylate copolymer onto which has been grafted acrylic acid (German Offenlegungsschrift 24 54 002), with a polyethylene onto which has been grafted butyl acrylate or acrylic acid and butyl acrylate, with an ethylene/propylene copolymer onto which has been grafted 30 butyl acrylate or styrene and acrylonitrile (German Offenlegungsschrift 29 02 468) or with a multiplestep polymer based on butyl acrylate, methyl acrylate and ethyl acrylate as well as optionally styrene graft cross-linked by allyl methacrylate or butylene diacrylate (German Offenlegungsschrift 27 26 256).

The effect achieved for reinforced PTMT with respect to notched bar impact resistance is 35 however small, that is to say the improvement in notched bar impact resistance amounts only to about 35 1 to 3 units from 9 to 11 KJ/m^2 , and is accompanied by a significant drop in the mechanical and thermal properties and in particular a drop in both the stiffness and Youngs modulus values.

There is, in particular no teaching in prior art documents as to how impact resistance and notched bar impact resistance can be effectively improved with reinforced PTMT. The stabilisation of PTMT and its stability on working up is moreover insufficient, and the aforementioned drop in molecular weight as 40

From the foregoing, it will therefore be appreciated that there has existed a need to produce a result of degradation is very significant. PTMT-shaped bodies having high impact and notched bar impact resistance, especially when reinforced, whose usual properties are good, and which additionally undergoes no significant reduction in nominal molecular weight during working. Such bodies thus need to possess good material properties, in general. Moreover the impact resisting effect should be achieved with small additive

Since PTMT is combustible and imflammable, an increasing amount of reinforced and unreinforced PTMT needs to be rendered flame resistant. However the usual monomeric bromine-50 containing flame protective agents have been found to be not very suitable for this purpose.

The flame protecting agent must withstand temperatures of 240 to 280°C in the extrusion and injection moulding working. When working PTMT which contains a flame protecting agent, presumably because of the effect of shearing forces, degradation of the PTMT occurs so that the final injected bodies of PTMT are of significantly lower molecular weight than the starting PTMT employed for the 55 extrusion, as a result of which a product having inferior mechanical properties, especially lower

The mechanical properties are also unsatisfactory when $\mathrm{Sb_2O_3}$ is present as synergist for the strength, than expected results.

This deterioration of the mechanical properties has meant that it has not hitherto been possible to flame protecting agent. 60 produce an unreinforced PTMT which is both flame resistant and has satisfactory mechanical properties. In particular the impact resistance and above all the elongation at break were very strongly influenced. Thus for example commercially available PTMT products which contain flame protecting agents as aforesaid have an impact resistance so far that test samples show between 25% and 100% break in comparison to "not broken" for pure PTMT test samples and the elongation at break lies

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between 12% and 17% (in comparison with more than 180% for pure PTMT, the second process of the protecting agent). For reinforced PTMT the drop in mechanical strength in like manner without flame protecting agent). For reinforced PTMT the drop in mechanical strength in like manner without flame protecting agent is likewise very disadvantageous, there values caused by the presence of a flame protecting agent is likewise very disadvantageous, there values caused by the presence of a flame protecting agent is likewise very disadvantageous, there being a fall in the impact resistance from 36 to 25—30 KJ/m² and in notched bar impact resistance from 9 to 10 to 5—7 KJ/m² in contrast to pure PTMT.

from 9 to 10 to 5—7 KJ/m² in contrast to pure PIMI.

Moreover, because such flame protected PTMT-shaped bodies have a lower stability during working and use than reinforced PTMT or reinforced PTMT which is not flame protected there is much scope for problems in the event of for example a breakdown in the injection moulding working, too long residence in the injection moulding machine, unsuitable machine adjustment or use of temperatures residence in the injection moulding machine, unsuitable machine adjustment or use of temperatures residence in the injection moulding machine, unsuitable machine adjustment or use of temperatures are further reduction in molecular weight and products with further deteriorated mechanical properties are obtained.

obtained.

Such flame protected, but mechanically deteriorated products will generally be unusable. In the Such flame protected, but mechanically deteriorated products will generally be unusable. In the use test with storage of the extrudates for 3 days at 150°C in air it is observed that the impact use test with storage of the extrudates for 3 days at 150°C in air it is observed that the impact resistance to below 3 resistance can drop to below 50 KJ/m² at 100% break, the notched bar impact resistance to below 3 resistance can drop to below 50 KJ/m² at 100% break, the notched bar impact resistance to below 3 solutions.

is obtained under the test conditions. It has previously been proposed to stem the reduction of the molecular weight degradation by PTMT-shaped bodies by adding to the PTMT a polymeric epoxy resin of equivalent weight 300 to 2000 PTMT-shaped bodies by adding to the PTMT a polymeric epoxy resin of equivalent weight 300 to 2000 PTMT-shaped bodies by adding to the PTMT a polymeric epoxy resin of equivalent weight 300 to 2000 PTMT-shaped bodies by adding to the PTMT and Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way there is g/mol in amounts between 0.5 and 20% (German Offenlegungsschrift 28 21 292). In this way the companies of the companies of the companies of the compa

$$(\eta_{\rm red} = \frac{\eta - \eta}{\eta} \cdot 1/c$$

wherein η =viscosity of the solvent phenol/o-Dichlorobenzene=60/40, η =viscosity of the solution and c=concentration of the solution in α /100 ml).

25 c=concentration of the solution in g/100 ml).
Moreover, the suitability for use, the results obtained in the use test carried out on the final extrudate and the η_{red} value in the extrudate also remain unsatisfactory if, according to German Offenlegungsschrift No. P 28 21 292, resins, PTMT, polypentabromobenzyl acylate and Sb₂O₃ are employed together (see Table 6, Comparative Examples H and J).

There exists therefore the need also to provide conveniently an unreinforced or reinforced PTMT which contains only a small amount of flame protecting agent and of impact resisting agent of the which contains only a small amount of flame protecting agent and injection moulding aforementioned co- or graft polymeric type, and which after extrusion and injection moulding possesses comparable mechanical and special properties to injection moulded products formed of pure possesses comparable mechanical and special properties as additives.

More particularly, in general, there is a need to be able to provide PTMT simultaneously with a number of special types of additives to impart reinforcement, impact resistance and flame protection to number of special types of additives to impart reinforcement, impact resistance and flame protection to number of special types of additives to impart reinforcement, impact resistance and flame protection to number of special types of additives are incorporated therein.

While avoiding the degradation or strong deterioration of the good material properties of PTMT it, while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT it, while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of PTMT while avoiding the degradation or strong deterioration of the good material properties of the good materia

which usually takes place when additives are incorporated therein.

According to the present invention, there is provided a polytetramethyleneterephthalate shaped

40 body which is formed of polytetramethylene-terephthalate and contains triglycidyl isocyanurate and/or

a bisovazoline as stabiliser.

a bisoxazoline as stabiliser.

It has been found the use of quite specific stabilisers, namely triglycidyl isocyanurate and/or a lt has been found the use of quite specific stabilisers, namely triglycidyl isocyanurate and/or a bisoxazoline, for example the phenylene or tetramethylene isoxazoline leads to suppression of the aforementioned undesirable phenomena. The effect of these stabilisers is particularly surprising since stabilisers of other type do not hinder the degradation of PTMT and do not lead to maintenance of the special material properties of PTMT.

special material properties of PTMT.

The present invention will now be described in detail with reference to four types of additives

The present invention will now be described in detail with reference to four types of additives

which may be present in polytetramethylene terephthalate shaped bodies, namely (1) reinforcing fillers,

which may be used alone or in combination

50 which may be used alone or in combination.

Thus this invention is applicable, not only to unreinforced PTMT shaped bodies but to reinforced PTMT-shaped bodies, especially such bodies when reinforced with glass fibres. Preferably from 15 to 80% by weight of reinforcing fillers, related to PTMT are present.

80% by weight of reinforcing fillers, related to PTMT are present.

Glass fibres have the advantage of possessing a so-called active reinforcing effect. Otherwise it is possible to use, for example asbestos fibres or organic polymer fibres, for example high melting polyester, polyurethane, polysulphone, aliphatic or aromatic polyamide fibres as reinforcing fillers.

Moreover this invention provides shaped polytetramethylene terephthalate bodies with contents of co or graft polymers of ethylene and vinyl acetate or co- or graft polymers based on acrylic acid or methacrylic acid esters as impact resisting agents. The impact resisting agents in the form of the co- or graft polymers are preferably added in amounts of 5 to 30, more preferably 12 to 25% by weight related to PTMT.

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otecting agents based This invention also provides PTMT shaped bodies which contain flame on polymerised bromine-containing acrylates or methacrylates and optionally also the aforementioned

The stabilisers surprisingly cause the molecular weight drop on working in the extruder and in the 5 subsequent injection mould to be reduced very much; even when using small amounts of the additives, can the degradation of PTMT be suitably influenced. The largely hindered degradation of PTMT on use of the shaped bodies is valuable. The hydrolysis of PTMT on standing in the ambient atmosphere is almost completely held back. The impact resisting agents are only effective as a result of the stabilisers added according to the invention. Moreover, as a result of the presence of the stabilisers flame protecting agents based on high polymerised bromine-containing acrylates which are solid materials,

The addition of ethylene/vinyl acetate co- or graft polymers is especially valuable taken together lead to only slight degradation. with reinforcing fillers. Of especial value are such PTMT bodies reinforced with glass fibres consisting of:

a) 45—83, preferably 55—71 parts by weight PTMT with a reduced specific viscosity $\eta_{\rm red}$ of 1.2—2.2 dl/g preferably 1.5—1.9 dl/g, ($\eta_{\rm red}$ is defined as previously indicated).

b) 15—40, preferably 25—35 parts by weight glass fibres,

c) 2-20, preferably 4-10 parts by weight of ethylene/vinyl acetate copolymers or graft d) 3-30, preferably 10-20% by weight of co- or graft polymers based on acrylic acid esters or

Furthermore, unreinforced shaped bodies can also be produced, containing 85 to 98% by weight 20 methacrylic acid esters.

PTMT and 2 to 15% by weight EVA and/or 3 to 30% by weight flame protecting agent which is a polymerised bromine-containing acrylate or methacrylate.

It is surprising that the aforementioned comparatively small amounts of EVA are sufficient. No further increased impact resistance can be achieved with larger amounts of EVA. 25

Suitable copolymers of ethylene and vinylacetate are those with contents of vinyl acetate of 2 to 65% by weight, preferably 5 to 50% by weight. The copolymers are known materials and can be produced by copolymerisation in suspension or in bulk. They are generally used in powder form.

By the terms co- or graft polymers based on acrylic acid esters or methacrylic acid esters are 30 understood those polymers in which at least 5% by weight, preferably from 10% by weight of an acrylic acid and/or methacrylic acid ester with 1 to 10, preferably 1 to 6 carbon atoms in the alcohol group of the ester, are present. The second component of the such polymers is preferably another of the indicated acrylic or methacrylic acid esters of a different type of polymerisable olefinic compound such 35 as for example an olefin, acrylonitrile or styrene. In the case of ternary polymers, there will be a further 35 one of the indicated monomers—in the case of graft polymers a graft substrate of two of the components or of butadiene-styrene and preferably an acrylic acid or methyacrylic acid ester is present as grafting agent. Preferred are mixed polymers of 20 to 50, preferably 30 to 40% by weight of an acrylic or methacrylic acid methyl or ethyl ester and a further acrylic or methacrylic acid propyl to butyl 40 ester as well as ter- and graft polymers of 5 to 40, preferably 10 to 30% by weight of an acrylic or 40 methacrylic acid ester, especially a methyl ester, and 40 to 80, preferably 50 to 70% by weight

The combined effect of the stabilisers and the impact resisting co- or graft polymers is surprising butadiene and 5 to 35, preferably 20 to 30% by weight styrene. for in the absence of the claimed stabilisers, co- or graft polymers (comparative examples A to G) 45 achieve practically no improved impact or notched bar impact resistance in comparison with the basic 45 material. In addition to improving the notched bar resistance, the addition of the stabilisers achieves an

Furthermore the stabilisers affect favourably the temperature stressing to which the material can increase in the elongation at break and the Youngs modulus. be subjected during working and increases in particular the working range, that it makes possible the 50 use of a greater temperature range in the extruder or in the injection moulding machine without harming and loss of the composition. Stabilisers of other type do not achieve this synergistic composite

The stabiliser(s) is/are preferably used in an amount of 0.1 to 2.5, more preferably 0.1 to 1.5% by weight based on the weight of PTMT. Insofar as triglycidyl isocyanurate is used alone, an upper limit of effect. 55 1.0% by weight is preferably placed on the amount added; insofar as p- or m-bisoxazoline is used, its quantity preferably amounts to not more than 1.5% by weight.

It is preferred to add as well a phosphitic antioxidant and a phenolic antioxidant additional to triglycidyl isocyanurate and/or the bisoxazoline. In general, it is feasible for only one of the two

Phosphitic antioxidants are phosphites of at least tri-functional alcohols, especially of pentaerythritol, or for example of trimethylolpropane or possibly even glycerol, wherein one or two of antioxidants to be added. the OH-groups of the phosphoric acid are esterified with a fatty alcohol residue, especially a C_8 to C_{24} fatty acid residue, preferably the stearyl residue or the decyl residue. The preferred alcohol is pentaerythritol.

Phenolic antioxidants will generally be of the type in which there is a sterically hindered phenolic

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H-group. Examples g a t-butyl group in the o-position to the pheno of such anti-oxidants are many. Preferred examples are, however, 2,2'-thiodiethylbis-[3-(3,5-ditert.butyl-4-hydroxyphenyl)]-propionate and 1,3,5-trimethyl-2,4,6-tri-(4-hydroxy-3,5-di-

Triglycidyl isocyanurate and one of the bisoxazolines achieves, alone or together, a decisive tert.butyl)benzene. improvement in the mechanical properties, although the added amount is small. The reduced viscosity $\eta_{
m red}$ of the PTMT in the injection mould after extrusion and working is surprisingly equally high or even higher than that of the pure PTMT starting material.

Moreover the two above-indicated types of antioxidant improve very much the scope for working, 10 the stability in use for example as measured in the indicated use test, as well as the colour of the 10 product; that is discolouration is avoided.

In general, when using antioxidants in the production of shaped bodies, it is preferred to use from 0.02 to 0.5, more preferably 0.05 to 0.3% by weight 2,2'-thiodiethyl-bis-[3-(3,5-ditert.butyl-4hydroxyphenyl)]-propionate and/or 0.05 to 0.5, more preferably 0.1 to 0.3% by weight distearyl pentaerythritol phosphite, especially with the aforementioned amounts of the stabilisers. When an 15 impact-resisting agent and/or a flame protecting agent is present the amount of the 2,2'-thiodiethylbis-[3-(3,5-ditert.butyl-4-hydroxyphenyl)]-propionate is preferably 0.02 to 0.3, more preferably 0.05 to 0.2% by weight. These percentages are based on the weight of PTMT.

In the production of reinforced PTMT use is preferably made of PTMT with $\eta_{\rm red}$ =1.2 to 1.3, since 20 here a PTMT of η_{red} =1.6 to 1.7, which is not only further condensed, but is also more expensive, brings no essential advantage. For unreinforced PTMT, in contrast, PTMT of $\eta_{\rm red}$ =1.6 and higher is preferred.

Reference has already been made herein to the application of this invention to unreinforced or reinforced PTMT-shaped bodies containing flame protecting agents which are polymerised bromine-

While some acceptable losses in impact resistance, notched impact resistance and elongation at containing acrylates or methacrylates. break, are obtained when using such flame protecting agents, it is possible as a result of stabilisation according to the invention to produce a satisfactory unreinforced flame protected PTMT based on PTMT—1.2, that is $\eta_{\rm red}$ =1.2 (Examples 21 and 22) when, without stabilisation or with addition of bisphenol-A-diglycidyl ethers, a complete injection moulding is obtained which is a completely 30 unusable material for constructional uses.

As polymeric, bromine-containing acrylates, there can be used homo- and copolymers containing units of the formula

wherein X signifies preferably bromine, optionally with proportions of chlorine, p the degree of 35 polymerisation, n=0 or 1, Y denotes Br or —CH₃ and R hydrogen, methyl, ethyl or propyl. The 35 production of such polymers is known from German Auslegesschrift 25 27 802. It is also possible to use polymers of such type obtained from the monomers of German Offenlegungsschrift 28 00 020, polymers and copolymers according to German Offenlegungsschrift 26 12 843 as well as copolymers according to German Offenlegungsschrift 26 48 969, and the corresponding bromine-containing 40 acrylates of benzylic alcohols or xylylene glycols. It is preferred, however, to use the especially simply 40 preparable PBB-PA, that is polymers of pentabromobenzyl acrylate, optionally with proportions of tribromo or tetrabromobenzyl acrylates. In addition to the aforementioned acrylates, the corresponding methacrylates can be used, on the basis of comparability, if where in the examples PBB-PA is given as a flame protective agent, a correspondingly good flame protecting effect is achievable with the other 45 known bromine-containing polymeric acrylates, especially if the bromine content approximately equals 45 that of PBB-PA. The flame protecting agent is preferably used in amounts of 5 to 20% by weight related PTMT.

 $\mathrm{Sb_2O_3}$ can be used as synergist to the flame protecting agent in amounts of from 1 to 10% by

In testing for the extent of flame protection achieved, as in the examples which follow, the 1.6 weight related to PTMT. mm flat rods required according to UL 94 (Underwriters Laboratories, subject UL 94) and LOI (lowest oxygen index, that is the O_2 proportion in an O_2/N_2 mixture in volume % which acts just selfextinguishingly) are produced on an injection moulding machine made by the ARBURG COMPANY.

The general mechanical properties were determined according to the following test 55 specifications:

discolouration of the product are therefore largely reduced.

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In particular, the reternant of stability in use as a result of the stabilisation system has been shown in a test in air at 180°C. This test shows that the impact resistance is maintained at "not broken" with unstabilised PTMT for only a few days, whereas with stabilised samples after 10 days the impact resistance has the value not broken and the notched bar impact resistance is likewise reduced 5 only slightly after about 10 days. In a similar manner, a PTMT shaped body reinforced with glass fibres exhibits only a small reduction in the impact resistance and notched bar impact resistance when subjected over many days to thermal stressing.

A further important effect is the stabilisation with respect to hydrolysis which can be checked by hot water storage of injection mouldings at 80°C. Here, the test shows that the original impact 10 resistance, notched bar impact resistance and elongation at break with unstabilised samples is only maintained for about 4 weeks, whereas the original values of impact resistance and notched bar impact resistance are maintained for about 12 weeks in the hydrolysis test carried out on the stabilised samples. This applies in particular for unreinforced PTMT; the effect of stabilisation of PTMT with glass fibre reinforcement is not quite so impressive.

The following Examples illustrate this invention. In the examples all parts are on a weight basis.

In each of these examples, and the associated Comparative Examples, to produce the indicated Examples 1 to 22 PTMT composition, the dry components were premixed well and co-extruded through a single screw extruder with a screw diameter of 45 mm in the temperature range 240 to 280°C. The extruded strand 20 was withdrawn through a water bath and granulated. After the drying of the granulate, the injection mouldings required for the testing of the mechanical and thermal properties were produced on an injection moulding machine of the firm KRAUSS-MAFFEI in a universal mould.

Examples 1 to 3 and Comparative Example A use ethylene-vinyl acetate copolymer (EVA) with 18% by weight vinyl acetate, Examples 4 and 5 as well as Comparative Examples B to D the same but 25 with 7.5% by weight vinyl acetate. GF denotes glass fibre. The impact resisting graft polymer M_1 consists of about 20% by weight methylmethacrylate, about 60% by weight butadiene and about 20% by weight styrene. the mixed polymer M₂ consists of about 65% by weight n-butyl acrylate and about 35% by weight methyl methacrylate.

Moreover, in the Examples, the reference letters I) to VII) have the following means:

- 30 I) 2,2'-Thiodiethylbis-3-(3,5-ditert.butyl-4-hydroxyphenyl)-propionate 30
 - II) Distearyl pentaerythrityl phosphite
 - III) Triglycidyl isocyanurate
 - IV) p-phenylenebisoxazoline
 - V) $\eta_{\rm red} = \eta_{\rm sp/c}$ (1% in Phenol/O-Dichlorobenzene=60/40)
- VI) Araldite GY 250 CIBA-GEIGY (Epoxide-equivalent weight about 180—190 g/mol Epoxide) 35 VII) Epikote 1001 of SHELL COMPANY

The PTMT samples used in the examples are denoted by reference letters as follows:

For PTMT-A,
$$\eta_{red}$$
=1.76 dl/g
PTMT-B, η_{red} =1.67 dl/g
40 PTMT-C, η_{red} =1.6 dl/g
PTMT-D, η_{red} =1.2 dl/g

Other abbreviations appearing in the examples are B=broken NB=not broken

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|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------|
| 5 | Table 1 Example 1 64 parts PTMT-A 30 parts GF 6 parts EVA Stabiliser: | | Example 2 60 parts PTMT-A 30 parts GF 10 parts EVA Stabiliser: 0,2 part I) | Comparative Example A 64 parts PTMT-A 30 parts GF 6 parts EVA |
| | | 0.1 part l) +0.2 part ll) | +0.2 part II) | without Stabiliser |
| | • | +0.4 part III) | +0.5 part III) | 36 |
| . D. Jacones | KJ/m² | 46 | 49 | |
| 10 Impact Resistance Notched bar impact resistance: | | 0.4 | 25 | 14 |
| Notched bar impact resistance | KJ/m² | 24 | 20 | 11 |
| 23°C | KJ/m² | 18 | 18 | 10 |
| _20°C | KJ/m² | 17 | 119 | 110 |
| -40°C | N/mm² | 124 | | 7000 |
| 15 Tear resistance | _ | 0.400 | 8800 | 7600 |
| Youngs Modulus (Drawing | N/mm² | 9400 | 182 | 173 |
| test) Limit of bending test | N/mm² | 190 | | |
| Thermal non-deformability | | | | 470 |
| 49 4 | | 186 | 178 | 172 >200 |
| 20 according to ISO/R75: A | ۰Č | >200 | >200 | >200 |
| ISO/R75: B | °C | 151 | | _ |
| Ball pressure hardness | N/mm² | 101 | | |
| Ball pressure transmission | | | | |
| | Table | 1 (continued) | | Example 5 |
| 25 | 102.0 | Example 3 | Example 4 | 60 parts PTMT-A |
| 23 | | LAGINAT A | CA morte PTMI-A | OU parter |
| | 4 | se narts PIMI-A | 64 parts PTMT-A | ลก narts GF |
| | (| 66 parts PTMT-A | 30 parts GF | 30 parts GF |
| | ; | 30 parts GF | 30 parts GF 6 parts EVA | 30 parts GF 10 parts EVA |
| | • | 30 parts GF 4 parts EVA | 30 parts GF 6 parts EVA Stabiliser: | 30 parts GF 10 parts EVA Stabiliser: |
| 30 | • | 30 parts GF 4 parts EVA Stabiliser: 0.1 part I) | 30 parts GF 6 parts EVA Stabiliser: 0,1 part I) | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part II) |
| 30 | • | 30 parts GF 4 parts EVA Stabiliser: 0.1 part I) +0.2 part II) | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part II) |
| 30 | • | 30 parts GF 4 parts EVA Stabiliser: 0.1 part I) +0.2 part II) | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part III) +0.4 part III) | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) |
| | | 30 parts GF 4 parts EVA Stabiliser: 0.1 part I) | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part II) +0.4 part III) 48 |
| Lunget Resistance | • | 30 parts GF 4 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.6 part III) | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 |
| Impact Resistance 35 Notched bar impact resistance: | KJ/m² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part I) +0.2 part III) +0.6 part IIII) 44 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 47 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 |
| Impact Resistance 35 Notched bar impact resistance: 23°C | KJ/m² KJ/m² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II +0.2 part III) +0.6 part IIII 44 19 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 23 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C | KJ/m² KJ/m² KJ/m² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II +0.2 part III) +0.6 part IIII 44 19 16 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part) +0.2 part) +0.4 part) 47 23 17 15 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 |
| Impact Resistance 35 Notched bar impact resistance: 23°C | KJ/m² KJ/m² KJ/m² KJ/m² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II +0.2 part III +0.6 part IIII 44 19 16 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 23 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C -40°C Tear resistance | KJ/m² KJ/m² KJ/m² KJ/m² N/mm² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II +0.2 part III) +0.6 part IIII) 44 19 16 14 129 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 47 23 17 15 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 115 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C -40°C Tear resistance 40 Youngs Modulus (Drawing | KJ/m² KJ/m² KJ/m² KJ/m² N/mm² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part III +0.2 part IIII +0.6 part IIII 19 16 14 129 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part) +0.2 part) +0.4 part) 47 23 17 15 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C -40°C Tear resistance 40 Youngs Modulus (Drawing test) | KJ/m² KJ/m² KJ/m² KJ/m² N/mm² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part III +0.2 part IIII +0.6 part IIII 19 16 14 129 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 47 23 17 15 126 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 115 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C -40°C Tear resistance 40 Youngs Modulus (Drawing test) | KJ/m² KJ/m² KJ/m² KJ/m² N/mm² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II +0.2 part III) +0.6 part IIII 44 19 16 14 129 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 47 23 17 15 126 9700 196 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 115 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C -40°C Tear resistance 40 Youngs Modulus (Drawing test) Limit of bending test Thermal non-deformability | KJ/m² KJ/m² KJ/m² KJ/m² N/mm² N/mm² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II) +0.2 part III) +0.6 part IIII 19 16 14 129 9800 187 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 47 23 17 15 126 9700 196 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 115 8600 182 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C -40°C Tear resistance 40 Youngs Modulus (Drawing test) Limit of bending test Thermal non-deformability according to | KJ/m² KJ/m² KJ/m² KJ/m² N/mm² N/mm² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II) +0.2 part III) +0.6 part IIII) 44 19 16 14 129 9800 187 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 47 23 17 15 126 9700 196 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 115 8600 182 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C -40°C Tear resistance 40 Youngs Modulus (Drawing test) Limit of bending test Thermal non-deformability according to 45 ISO/R75: A | KJ/m² KJ/m² KJ/m² KJ/m² N/mm² N/mm² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II +0.2 part III) +0.6 part IIII) 44 19 16 14 129 9800 187 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 47 23 17 15 126 9700 196 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 115 8600 182 |
| Impact Resistance 35 Notched bar impact resistance: 23°C -20°C -40°C Tear resistance 40 Youngs Modulus (Drawing test) Limit of bending test Thermal non-deformability according to | KJ/m² KJ/m² KJ/m² KJ/m² N/mm² N/mm² | 30 parts GF 4 parts EVA Stabiliser: 0.1 part II +0.2 part III) +0.6 part IIII) 44 19 16 14 129 9800 187 | 30 parts GF 6 parts EVA Stabiliser: 0.1 part I) +0.2 part II) +0.4 part III) 47 23 17 15 126 9700 196 | 30 parts GF 10 parts EVA Stabiliser: 0.2 part I) +0.3 part III) +0.4 part IIII) 48 26 18 17 115 8600 182 |

| | Table 1 (| continued) | | Comparative |
|-------------------------------------------------------------------------------------------------------|-------------------------------------------|----------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------|
| 5 | 60 30 | Comparative Example B parts PTMT-A parts GF parts EVA Without stabiliser | Comparative Example C 70 parts PTMT-A 30 parts GF Without stabiliser | Example D 70 parts PTMT-A 30 parts GF Stabiliser: 0.1 part I) +0.2 part III) +0.5 part III) |
| 10 Impact Resistance | KJ/m² | 38 | 35 | |
| Notched bar impact resistance. 23°C —20°C —40°C | KJ/m² KJ/m² KJ/m² N/mm² | 13 12 11 112 | 9—10 — 8—9 130 | 9—11 — 8—10 133 |
| 15 Tear resistance Youngs Modulus (Drawing test) Limit of bending test Thermal non-deformability | N/mm² N/mm² | 6700 164 | 10000 190 | 10500 200 |
| 20 according to ISO/R75: A ISO/R75: B Ball pressure hardness | °C °C N/mm² | 168 >200 — | 200 >200 175 | _ _ _ |
| 25 30 | . | Table 2 Example 6 55 parts PTMT-A 15 parts M ₁ Stabiliser: 0.1 part I) +0.2 part III) without break | Example 7 88 parts PTMT-A 12 parts M ₁ Stabiliser: 0.1 part I) +0.2 part III) +0.6 part III) without break | |
| Impact resistance Notched bar impact resistance: 23°C 35 —20°C —40°C Tensile strength | KJ/m² KJ/m² KJ/m² N/mm² N/mm² | 30 | 32 14 11 45 32 107 | 47 19 16 42 32 140 |
| Resistance to tear Elongation at break 40 Bending resistance Youngs Modulus (from bending experiment) | % N/mm² N/mm² | 125 69 | 72 1980 | 65 1850 |
| Thermal non-deformability according to ISO/R75: A ISO/R75: B | °C °C | 55 151 | 56 154 | 51 143 |

| | | Table 2 (cor | ntinued) | | |
|----|----------------------------------------|----------------|----------------------------------------------------------------|-------------------------------------------------------------------------|------|
| | | 85 15 | Example 9 parts PTMT-A parts M ₁ abiliser: | Example 10 82 parts PTMT-A 18 parts M ₁ Stabiliser: | 5 |
| 5 | | | 0.1 part I) +0.2 part II) +0.4 part IV) without break | 0.1 part I) +0.2 part II) +0.5 part IV) without break | 10 |
| | Impact resistance | KJ/m² | Williode Disco | 40 | 10 |
| 10 | Notched bar impact resistance. 23°C | KJ/m² KJ/m² | 45 17 | 48 | |
| | -20°C | KJ/m² | 13 44 | 40 | 4 == |
| | -40°C Tensile strength | N/mm² | 30 | 29 | 15 |
| | Resistance to tear | N/mm² | 130 | 115 | |
| 15 | Elongation at break | % N/mm² | 64 | 61 | |
| | Rending resistance | Milin | | 1700 | |
| | Vounds Modulus (from | N/mm² | 1750 | 1700 | 20 |
| | banding experiment/ | | • | | |
| 20 | Thermal non-deformability | | | | |
| | according to | °C | _ | | |
| | ISO/R75: A ISO/R75: B | °C | | | |
| | | Table 2 (c | ontinued) | | 25 |
| 25 | | 10010 = 10 | Comparative Example E | Comparative Example F 82 parts PTMT-A | 20 |
| 25 | | | 85 parts PTMT-A | 18 parts M ₁ | |
| | | | 15 parts M ₁ | without | |
| | | | without Stabiliser | Stabiliser | 30 |
| | | | without break | k without break | |
| 30 | Impact resistance | KJ/m² | Williods | | |
| | Notched bar impact resistance: | KJ/m² | 12 | 16 | |
| | 23°C | KJ/m² | 8 | 10 7 | 35 |
| | _20°C | KJ/m² | ь | , 39 | |
| 35 | 40°C | N/mm | 2 42 | 27 | |
| | Tensile strength | N/mm | 2 28 | 30 | |
| | Resistance to tear | % | 35 | 55 | 40 |
| | Elongation at break | N/mm |) ² 5/ | | 40 |
| | Bending resistance | | 4.500 | 1450 | |

N/mm²

°C

145

-40°C
Tensile strength
Resistance to tear
Elongation at break
Bending resistance
Youngs Modulus (from
bending experiment)
Thermal non-deformability
according to
ISO/R75: A

| · · | | | | | |
|-----|---------------------------------------------------|--------------|----------------------|-------------------------|-----|
| | | Table 3 | | | |
| | | | xample 11 | Example 12 | |
| | | OF = | arts PTMT-B | 80 parts PTMT-B | |
| | | | | 20 parts M ₂ | |
| | | 10 [| parts M ₂ | Stabiliser: | 5 |
| - | | | biliser: | 0.1 part) | • |
| 5 | | | 0.1 part l) | +0.2 part II) | |
| | | | -0.2 part II) | +0.6 part IV) | |
| | | 4 | -0.4 part IV) | without break | |
| | | KJ/m² | without break | Without preak | 10 |
| | Impact resistance | · | | 0.0 | . • |
| 0 | Notched bar impact resistance: | KJ/m² | 23 | 26 | |
| | 23°C | KJ/m² | 9 | 11 . | |
| | _20°C | KJ/m² | 7 | 8 | |
| | _40°C | | 43 | 41 | |
| | Tensile strength | N/mm² | 29 | 28 | 15 |
| 15 | Resistance to tear | N/mm² | 60 60 | 65 | |
| , | Elongation at break | % | 70 | 57 | |
| | Bending resistance | N/mm² | 70 | • | |
| | Youngs Modulus (from | | 4050 | 1850 | |
| | bending experiment) | N/mm² | 1950 | ,500 | 20 |
| | Thermal non-deformability | | | | |
| 20 | Inermal non-determation | | | EO | |
| | according to | °C | 54 | 52 4.27 | |
| | ISO/R75: A | ٥Č | 148 | 137 | |
| | ISO/R75: B | J | | | |
| | | Table 3 (con | tinued) | | 25 |
| 25 | | • | Example 13 | O | 29 |
| 25 | | 8 | 5 parts PTMT-B | Comparative | |
| | | 1 | 5 parts M₂ | Example G | |
| | | S | tabiliser: | 85 parts PTMT-B | |
| | | J | 0.2 part I) | 15 parts M ₂ | |
| | | | +0.2 part II) | without | 30 |
| 30 | | | +0.5 part III) | stabiliser | |
| | | · // ·/2 | without break | without break | |
| | Impact resistance | KJ/m² | AAITIIOGE DIOGIC | | |
| | Notched bar impact resistance: | | 21 | 14 | |
| | 23°C | KJ/m² | ۷1 | - | 35 |
| 35 | _20°C | KJ/m² | . | 5 | - |
| 00 | -40°C | KJ/m² | | 44 | |
| | | N/mm² | 44 | 29 | |
| | Tensile strength | N/mm² | 30 | | |
| | Resistance to tear | % | 50 | 26 56 | 40 |
| | Elongation at break | N/mm² | 71 | 56 | 40 |
| 40 | Bending resistance | 14 | | | |
| | Youngs Modulus (from | N/mm² | 1970 | 1800 | |
| | bending experiment) | (W) HALL | ••• | | |
| | | | | | |
| | Thermal non-deformability | | | | |
| | Thermal non-deformability according to | 22 | | 55 | 45 |
| 45 | Thermal non-deformability according to ISO/R75: A | °C | | 55 136 | 45 |

. . .

Table 4 Unreinforced PTMT-compositions Example 14

| 5 | | 1 | Example 14 6 parts PTMT-C 0 parts PBB-PA 4 Sb ₂ O ₃ Stabiliser: 0.1 part I) +0.2 part II) +0.5 part III) | Example 15 86 parts PTMT-C 10 parts PBB-PA 4 parts Sb ₂ O ₃ Stabiliser: 0.1 part I) +0.1 part III) | Example 16 86 parts PTMT-C 10 parts PBB-PA 4 parts Sb ₂ O ₃ Stabiliser: 0.1 part I) +0.2 part II) +1.0 part IV) all NB |
|----|--------------------------------------------------------------------------|----------------|--------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------|
| 10 | | | all NB | all NB | 6.0 |
| | Impact resistance Notched bar impact resistance Bending resistance | KJ/m² N/mm² | 5.8 95 | 5.4 93 | 96 |
| | Youngs Modulus (from | N/mm² | 2600 | 2500 | 2600 |
| 15 | bending test) | N/mm² | 59 | 60 | 59 33 |
| | Tensile strength Tear resistance Elongation at break | N/mm² % | 34 120—140 | 37 40—60 | 100—120 |
| | Thermal non-deformability | | | | |
| 20 | according to | °C | 68 | 62 | 67 168 |
| | ISO/R75: A | ۰č | 167 | 161 | 100 |
| | ISO/R75: B Flame protection | | . VO | vo | VO |
| | according to UL 94 | % | 3031 | 30—31 | 30—31 1.82 |
| 25 | η_{red} (V) (Granulate) | | 1.81 | 1.76 | |
| | $\eta_{\rm red}$ (V) (injection | | 1.87 | 1.69 | 1.78 |
| | moulding) | | ••• | | |

Table 4 (continued) Unreinforced PTMT-compositions

| | Unreinforced PTM1 | r-composition | S | - 1-10 |
|-------------------------------------------------------------------------------------------------------------|--------------------------------------------|-------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------|
| 30 | Exa 86 pa 10 pai 4 par Stabili | nmple 17 rts PTMT-C rts PBB-PA rts Sb ₂ O ₃ iser: | Example 18 86 parts PTMT-D 10 parts PBB-PA 4 parts Sb ₂ O ₃ Stabiliser: 0.1 part I) | Example 19 86 parts PTMT-D 10 parts PBB-PA 4 parts Sb ₂ O ₃ Stabiliser: 0.1 part I) |
| Impact resistance | +0 +0 | .1 part I) .1 part II) .7 part IV) all NB 5.2 | +0.2 part II) +0.5 part III) 8NB:2B 4.0 | +0.2 part II) +1.2 part IV) 6NB:4B 3.8 98 |
| 40 Notched bar impact resistance Bending resistance Youngs Modulus (from | N/mm² N/mm² | 92 2500 | 94 2700 62 | 2800 61 |
| bending test) Tensile strength 45 Tear resistance Elongation at break | N/mm² N/mm² % | 61 34 60—80 | 56 10—20 | 60 10—20 |
| Thermal non-deformability according to ISO/R75: A 50 ISO/R75: B | °C °C | 59 163 | 57 164 | 58 162 |
| Flame protection according to UL 94 | % | VO 30—31 1.77 | VO 30—31 1.21 | VO 3031 1.28 |
| $oldsymbol{\eta}_{red}$ (V) (Granulate) 55 ${}^{\circ}oldsymbol{\eta}_{red}$ (V) (Injection moulding) | | 1.69 | 1.23 | 1.26 |

;

50

not resistant

1.76

1.67

| • | | | | GB 2 098 231 A | 12 |
|----|------------------------------------------------------------------------|----------------|---------------------------------------------------|----------------------------------------|----|
| 2 | | | | | |
| | • | Table 4 (con | tinued) | | |
| | Unrein | forced PTM1 | -compositions | O marathro | |
| | | | Comparative | Comparative Example J | |
| | | _ | Example H | 86 parts PTMT-C | 5 |
| 5 | | 80 | 6 parts PTMT-C | 10 parts PBB-PA | • |
| • | | 70 | 0 parts PBB-PA 4 parts Sb ₂ O₃ | 4 parts Sb ₂ O ₃ | |
| | | • | 4 parts 30 ₂ 0 ₃ Without | Stabiliser: | |
| | | | stabiliser | 3 parts V) | |
| | | | 8NB:2B | 8NB:2B | 10 |
| 0 | Impact resistance | KJ/m² | 4.1 | 4.0 | |
| | Notched bar impact resistance: | N/mm² | 93 | 95 | |
| | Bending resistance Youngs Modulus (from | , | | | |
| | bending test) | N/mm² | 2600 | 2500 | 15 |
| E | Tensile strength | N/mm² | 58 | 62 | 10 |
| 5 | Tear resistance | N/mm² | 32 | 36 10 - 30 | |
| | Elongation at break | % | 1020 | 10—20 | |
| | Thermal non-deformability | | | | |
| | according to | 00 | 61 | _ | 20 |
| 20 | ISO/R75: A | °C °C | 165 | | |
| | ISO/R75: B | -0 | 103 | | |
| | Flame protection | | VO | VO | |
| | according to UL 94 | % | 30—31 | 30—31 | 65 |
| | η'' LOI η_{red} (V) (Granulate) | /6 | 1.65 | 1.68 | 25 |
| 25 | η_{red} (V) (Grandiate) η_{red} (V) (Injection | | | | |
| | η _{red} (V) (Injection moulding) | | 1.54 | 1.58 | |
| | modiag/ | | | | |
| | l Inc | Table 4 | (continued) TMT-compositior | IS | |
| | Oil | i Giriloroca : | Comparative | | 30 |
| 30 | | | Example K | | |
| | | | 86 parts PTMT-C | | |
| | | | 10 parts PBB-PA | | |
| | | | 4 parts Sb ₂ O ₃ | Comparative | 35 |
| 35 | | | Stabiliser: | Example L | 35 |
| JU | • | | 4 parts VII) | <i>Pure PTMT-C</i> all NB | |
| | Impact resistance | 4 | 9NB:1B | 5.5 | |
| | Notched bar impact resistance: | KJ/m² | 4.2 96 | 86 | |
| | Bending resistance | N/mm² | 30 | | 40 |
| 40 | Youngs Modulus (from | N/mm² | 2600 | 2400 | |
| | bending test) | N/mm² | | · 55 | |
| | Tensile strength | N/mm² | | 34 | |
| | Tear resistance Elongation at break | % | 1020 | 182 | 4- |
| 45 | Thermal non-deformability | •• | | | 45 |
| 45 | according to | | | 00 | |
| | ISO/R75: A | ۰C | _ | 66 170 | |
| | ISO/R75: B | °C | | 170 | |
| | Flame protection | | _ | u at annintemt | 50 |

VO

%

30---31

1.66

1.52

Flame protection according to UL 94

 $\eta_{\rm red}$ (V) (Granulate) $\eta_{\rm red}$ (V) (Injection moulding)

LOI

Table 5 GF-reinforced PTMT-compositions

| 5 | | Example 20 58 parts PTMT-D 30 parts GF 8 parts PBB-PA 4 parts Sb ₂ O ₃ Stabiliser: | Example 21 58 parts PTMT-D 30 parts GF 8 parts PBB-PA 4 parts Sb ₂ O ₃ Stabiliser: | Example 22 58 parts PTMT-D 30 parts GF 8 parts PBB-PA 4 parts Sb_2O_3 Stabiliser: 0.1 part I) |
|----------------------------------------------------------------------------------------------------------------|-------------------------|----------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------|
| Impact resistance Notched bar impact resistance: Bending resistance Tensile strength Thermal non-deformability | KJ/m² KJ/m² N/mm² | 0.1 part I) +0.2 part II) +0.7 part III) 36 9—10 220 140 | 0.1 part I) +0.2 part II) +1.0 part IV) 33 910 210 135 | +0.1 part II) +0.5 part III) 35 10—11 220 145 |
| according to ISO/R75: A ISO/R75: B | °C °C | 198 >200 | | 194 >200 |
| 20 Flame protection according to UL 94 $^{\prime\prime}$ LOI $\eta_{\rm red}$ (V) (Granulate) | % | VO 29.530.5 1.31 | VO 1.31 | VO — 1.67 |
| η_{red} (V) (Injection 25 moulding) | • | 1.29 | 1.27 | 1.59 |

Table 5 (continued) GF-reinforced PTMT-compositions

| | • | , , , , , , , , , , , , , , , , , , , , | | | |
|----|--------------------------------------------------------------------------------------|-----------------------------------------|----------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------|
| 30 | | | Comparative Example M 58 parts PTMT-D 30 parts GF 8 parts PBB-PA 4 parts Sb ₂ O ₃ without stabiliser | Comparative Example N 58 parts PTMT-D 30 parts GF 8 parts PBB-PA 4 parts Sb ₂ O ₃ Stabiliser: 4 parts VI) | Comparative Example O 70 parts PTMT-D 30 parts GF |
| 35 | Impact resistance Notched bar impact resistance: Bending resistance Tensile strength | KJ/m² KJ/m² N/m² N/m² | 26 57 190 133 | 28 5—7 200 136 | 35 9—10 220 130 |
| 40 | Thermal non-deformability according to ISO/R75: A ISO/R75: B | °C °C | 186 >200 | 190 >200 | 200 >200 |
| 4- | Flame protection | | VO | VO | not resistant |
| 45 | according to UL 94 "LOI $\eta_{\rm red}$ (V) (Granulate) | % | 1.18 | 1.20 | 1.21 |
| | η_{red} (V) (Injection moulding) | | 0.94 | 0.98 | 1.06 |

10

15

20



Table 6 Change of impact resistance, notched bar impact resistance and elongation at break with time on storage of injection mouldings at 150°C

| 5 | Comparative Example J | Comparative Example H | Example 16 | xample 14 | E | |
|----|--------------------------|--------------------------|------------|-----------|------|--|
| | • | Impact resistance | | | Days | |
| | 8NB:2B | 8NB/2B | 10NB | 10NB | 0 | |
| | 4NB:6B | 10B | 10NB | 10NB | 3 | |
| | 10NB | 10B | 10NB | 10NB | 5 | |
| 10 | 10NB | 10B | 9NB:1B | 10NB | 7 | |
| 10 | 10NB | 10B | 9NB:1B | 8NB:2B | 10 | |
| | 10NB | 10B | 6NB:4B | 5NB:5B | 15 | |

Table 6 (continued)

| 15 | Comparative Example J tance | Comparative Example H ned bar impact resis | Example 16 Notch | Example 14 | Days |
|----|-----------------------------------|--------------------------------------------------|---------------------|------------|------|
| | 4.0 | 4.1 | 6.0 | 5.8 | 0 |
| | 3.5 | 3.1 | 4.7 | 5.4 | 3 |
| | 2.8 | 2.9 | 5.0 | 5.0 | 5 |
| 20 | 2.3 | 2.0 | 4.8 | 5.1 | 7 |
| | - | | 3.6 | 3.7 | 10 |
| | | | 3.4 | 3.6 | 15 |

Table 6 (continued)

| 25 | Days | Example 14 | Example 16 | Comparative Example H Elongation at break (%) | Comparative Example J | 25 |
|----|------|------------|------------|-----------------------------------------------------|--------------------------|----|
| | 0 | 125 | 110 | 12 | 15 | |
| | 3 | 94 | 70 | 9 | 11 | |
| | 5 | 70 | 58 | 5 | 6 | |
| 30 | 7 | 50 | 37 | 3 | 3 | 30 |
| | 10 | 40 | 30 | | | |
| | 15 | 36 | 26 | | _ | |

Example 23

An unreinforced PTMT ($\eta_{\rm red}$ =1.76 dl/g) was dusted with 0.1% (I), 0.2% (II) and 0.1% (III), the 35 percentages being based on the weight of PTMT, and extruded by co-extrusion through a single screw extruder with the temperature profile of 230 to 255°C. Then standard small rods were produced on the injection moulding machine. The standard small rods were stored under the air atmosphere at 180°C. The variation of impact resistance and notched bar impact resistance with time in comparison with unstabilised samples is shown from the following table.

Table 7

| | Impact resistance (KJ/m²) | | Notched bar impact resistance (KJ/m²) | |
|------|---------------------------|---------------|---------------------------------------|------------|
| Days | unstabilised | stabilised | unstabilised | stabilised |
| 0 | without break | without break | 5.1 | 5.5 |
| 1 | 28 | without break | 4.2 | 5.4 |
| 3 | | without break | 2.6 | 4.6 |
| 5 | | without break | 2.0 | 3.7 |
| 7 | _ | without break | 1.8 | 3.8 |
| 10 | | without break | _ | 4.2 |
| 15 | | 40 | | 4.0 |

In corresponding manner, PTMT with 30% by weight glass fibres and 0.1% by weight (I), 0.2% by weight (II) and 0.2% by weight (III), the percentages being based on the weight of PTMT, was provided and tested.

| - | . 1 | _ | 0 |
|----|-----|---|---|
| ıя | ומ | A | 0 |

| | Impact resistance (KJ/m²) | | Notched bar impact resistance (KJ/m²) | | | |
|------------------|---------------------------|----------------------|---------------------------------------|-----------------------|--|--|
| Days | unstabilised | stabilised | unstabilised | stabilised | | |
| 0 3 5 9 | 40 31 25 18 | 47 45 46 43 | 10 9 7 5.5 | 13 11 11 9.5 | | |

A corresponding PTMT with $\eta_{\rm red}$ =1.6 dl/g was stabilised against hydrolysis by addition of 0.1% by weight (I), 0.2% by weight (II) and 1.2% by weight (IV), based on the weight of PTMT and tested for resistance to hydrolysis by heat storage of the injection mouldings at 80°C.

This test showed that the original impact resistance after the working was retained in the hot water test for about 12 weeks, whereas the original impact resistance with unstabilised samples was only retained for 4, weeks. In addition, the good notched bar impact resistance and elongation values were maintained in the test for 10 to 12 weeks. This points to a trouble free behaviour in use. In contrast, the unstabilised samples only maintained satisfactory notched bar impact resistance and 10 elongation at break, which would allow their trouble-free use, for about 3 to 4 weeks.

Table 9

| | | | Tubio | | | |
|-------|------------------------------|---------------|------------------------------------------|------------|-------------------------|---------------------------------------|
| | Impact resistance (KJ/m²) | | Notched bar impact resistance (KJ/m²) | | Elongation at break (%) | |
| Weeks | unstabilised | stabilised | unstabilised | stabilised | unstabilised | stabilised |
| 0 | without break | without break | 5.2 | 6.0 | 190 | 225 |
| 2 | without break | without break | 4.8 | 5.9 | 30 | 136 |
| 4 | without break | without break | 4.5 | 5.4 | 13 | 80 |
| 6 | 90 | without break | 1.6 | 4.5 | 8 | 50 |
| 8 | 10 | without break | 0.9 | 4.8 | 1 | 34 |
| | 10 | without break | | 4.5 | | 24 |
| 10 | | without break | | 4.3 | _ | 18 |
| 12 | | | | 1.9 | _ | 16 |
| 14 | | 28 | | | | 12 |
| 16 | | 12 | | 1.4 | | · · · · · · · · · · · · · · · · · · · |

Claims

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- 1. A polytetramethylene-terephthalate shaped body which is formed of polytetramethyleneterephthalate and contains triglycidyl isocyanurate and/or a bisoxazoline as stabiliser.
 - 2. A body as claimed in Claim 1, in which the polytetramethylene terephthalate is reinforced.
- 3. A body as claimed in Claim 2, in which the polytetramethylene terephthalate is glass-fibre reinforced.
- 4. A body as claimed in Claim 2 or 3, which contains from 15 to 80% by weight of a reinforcing filler related to the polytetramethylene terephthalate.
- 5. A body as claimed in any one of the preceding claims, which additionally contains a phosphitic 20 20 anti-oxidant and/or a phenolic antioxidant.
 - 6. A body as claimed in Claim 5, wherein the phosphitic antioxidant is a phosphite of an at least trifunctional alcohol wherein one or two of the OH-groups of the phosphoric acid moiety are esterified with a fatty alcohol residue.
 - 7. A body as claimed in Claim 6, wherein the alcohol is pentaerythritol and the fatty alcohol contains from 8 to 24 carbon atoms.
 - 8. A body as claimed in Claim 7, wherein the alcohol is distearyl pentaerythritylphosphite.
- 9. A body as claimed in any one of Claims 5 to 8, wherein the phenolic antioxidant is 2,2'thiodiethylbis-[3-(3,5-di-tert.butyl-4-hydroxyphenyl)]-propionate or 1,3,5-trimethyl-2,4,6-tri-(4-30 hydroxy-3,5-di-tert.butyl)benzene.
 - 10. A body as claimed in any one of the preceding claims, which contains from 0.1 to 2.5% by weight, based on the amount of polymethylene terephthalate, of triglycidyl isocyanurate and/or bisoxazoline.
- 11. A body as claimed in Claim 10, which contains from 0.1 to 1.5% by weight, based on the 35 amount of polymethylene terephthalate, of triglycidyl isocyanurate and/or bisoxazoline.
 - 12. A body as claimed in Claim 10 or 11 which contains from 0.02 to 0.5% by weight of 2,2'thiodiethylbis[3-(3,5-ditert.butyl-4-hydroxyphenyl)]-propionate and/or from 0.05 to 0.5% by weight distearyl pentaerythritylphosphite.
 - 13. A horty as claimed in Claim 12, which contains from 0.05 to 0.3% by weight of 2,2'-

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thiodiethylbis[3-(3,5-ditert.baryl-4-hydroxyphenyl)]-propionate and/or from 0.3% by weight distearyl pentaerythritylphosphite. 14. A body as claimed in any one of Claims 1 to 11 which additionally contains a co- or a graft polymer based on an acrylic acid or methacrylic ester as impact-resisting agent. 15. A body as claimed in any one of Claims 1 to 11, which additionally contains a co-polymer of 5 5 ethylene and vinyl acetate as impact resisting agent. 16. A body as claimed in any one of Claims 1 to 11, which additionally contains a graft polymer of ethylene and vinyl acetate as impact resisting agent. 17. A body as claimed in Claim 15 or 16, in which the impact resisting agent contains from 2 to 10 10 65% by weight vinyl acetate. 18. A body as claimed in Claim 17, in which the impact resisting agent contains from 5 to 50% by weight vinyl acetate. 19. A body as claimed in Claim 14, wherein the impact resisting agent is a polymer of 20 to 50% by weight of an acrylic or methacrylic acid methyl or ethyl ester, together with additionally an acrylic or 15 15 methacrylic acid C₃ or C₄ alkyl ester. 20. A body as claimed in Claim 14, wherein the impact resisting agent is a polymer of 5 to 40% by weight of an acrylic or methacrylic acid ester, 40 to 80% by weight butadiene and 5 to 35% by weight styrene, the amounts of the respective monomers to total 100%. 21. A body as claimed in any one of Claims 14 to 20, which contains the impact resisting agent 20 20 in an amount of from 5 to 30% by weight of the polytetramethylene terephthalate. 22. A body as claimed in Claim 21, which contains the impact resisting agent in an amount of from 12 to 25% by weight of the polytetramethylene terephthalate. 23. A body as claimed in any one of Claims 1 to 11 and 14 to 22, which additionally contains a polymer of a bromine-containing acrylate or methacrylate as flame protecting agent. 24. A body as claimed in Claim 23, wherein the flame-protecting agent is a polymer of 25 25 pentabromobenzyl acrylate. 25. A body as claimed in Claim 23 or 24, which contains from 5 to 20% by weight of the flameprotecting agent, related to the weight of polytetramethylene terephthalate. 26. A body as claimed in Claim 24 or 25, which additionally contains Sb₂O₃ as synergist for the 30 30 flame protecting agent. 27. A body as claimed in Claim 26, which contains from 1 to 10% by weight of Sb₂O₃, related to the weight of polytetramethylene terephthalate. 28. A body as claimed in any one of claims 14 to 27, which contains from 0.02 to 0.3% by weight of 2,2'-thio-diethylbis[3-(3,5-ditert.butyl-4-hydroxy-phenyl)]-propionate and/or 0.05 to 0.5% 35 35 by weight of distearyl pentaerythritylphosphite. 29. A body as claimed in Claim 28, which contains from 0.05 to 0.2% by weight of 2,2'thiodiethylbis[3-(3,5-ditert.butyl-4-hydroxyphenyl)]-propionate and/or 0.1 to 0.3% by weight of distearyl pentaerythritylphosphite. 30. A polytetramethylene terephthalate shaped body, substantially as described in any one of the 40 40 foregoing Examples 1 to 5. 31. A polytetramethylene terephthalate shaped body, substantially as described in any one of the foregoing Examples 6 to 13. 32. A polytetramethylene terephthalate shaped body, substantially as described in any one of the foregoing Examples 13 to 22. 33. A polytetramethylene terephthalate shaped body as claimed in Claim 1, substantially as 45 45 described in the foregoing Example 23. 34. A process for the production of a stabilised polytetramethylene terephthalate shaped body as claimed in any one of Claims 1 to 29, which comprises continuously supplying to an extruder polytetramethylene terephthalate, triglycidyl isocyanurate and/or a bisoxazoline and optionally one or 50 more further components is/are chosen from a said reinforcing filler, said impact resisting agents and 50 said polymerised bromine-containing acrylate or methacrylate flame protecting agent, which component(s) is/are supplied separately from one another and separately from the polytetramethylene terephthalate, the triglycidyl isocyanurate and/or bisoxazoline and, if used, the phosphitic or phenolic antioxidant and/or Sb₂O₃ being simultaneously supplied continuously and separately from one another 55 to the extruder either together with the polytetramethylene terephthalate or one or more of said 55 components or as materials supplied separately from any other constituent(s) of the body, and coextruding said constituents in the extruder. 35. A process for the production of a stabilised polytetramethylene terephthalate shaped body,

substantially as described in any one of the foregoing Examples.